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# Roll-to-Roll Dyed Conducting Silk Yarns: A Versatile Material for E-Textile Devices

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Textiles are a promising base material for flexible and wearable electronic applications such as sensors, actuators, and energy harvesters. An essential component in such electronic textiles (e-textiles) is electrically conducting yarns. Here, a continuous dyeing process is presented to convert an off-the-shelf silk sewing thread into a wash and wear resistant functional thread with a conductivity of about  $70 \text{ S cm}^{-1}$ ; a record high value for coated yarns. An aqueous ink based on the conducting polymer:polyelectrolyte complex poly(3,4-ethylenedioxythiophene): polystyrene sulfonate (PEDOT:PSS) is modified, to produce more than 100 m of dyed conducting threads, which are subsequently converted into e-textiles by both hand weaving and machine embroidery. The yarns are resistant to abrasion and wear, and can be machine washed at least 15 times with retained electronic properties. The woven fabric is used to design a capacitive touch sensor which functions as an e-textile keyboard.

biocompatibility. The potential functionalities of e-textiles based on conjugated polymers include transistors,<sup>[1]</sup> data storage,<sup>[2]</sup> photovoltaics,<sup>[3]</sup> and thermoelectrics.<sup>[4]</sup> Electrically conducting fibers and yarns are a critical component in e-textiles,<sup>[5]</sup> where they constitute connectors, passive components, or building blocks for active components. Conducting fibers can be designed by several different principles including: fiber spinning of conjugated polymers, fiber spinning of blends, i.e., conjugated polymers combined with an insulating polymer, or dyeing and coating of existing fibers and fabrics.<sup>[6]</sup> The latter principle has the advantage that a base yarn or fabric can be selected with the desired form factor, mechanical properties, and processability, and then be functionalized through


## 1. Introduction

Flexible electronics promise to enrich our lives by adding sensor and logics functionality to everyday objects. For devices worn close to the body, electronic textiles (e-textiles) show great potential to complement or in themselves constitute wearable electronics. E-textiles may be developed from metallic materials, carbon allotropes (including carbon black, graphene, carbon nanotubes) or conjugated polymers. At present there exists considerable interest in using conjugated polymers for the manufacture of e-textiles, as such polymers display attractive mechanical properties, ease of processing as well as

the addition of conjugated polymers to the surface, with its original attractive properties retained. Coating and dyeing has been demonstrated using dip-coating<sup>[4b,7]</sup> and in situ polymerization either by wet chemistry<sup>[8]</sup> or employing oxidative chemical vapor deposition (oCVD).<sup>[9]</sup> In situ polymerization results in a thin conformal coating, which does not significantly affect the mechanical properties (flexibility, tensile strength, breathability) of the base textile. The thin coating layer, however, seems to limit the bulk conductivity to  $<1 \text{ S cm}^{-1}$ .<sup>[8a,d,g,9b]</sup> Dip-coating or exhaust dyeing, where the textile is functionalized by immersion in a dye-bath, is an accessible method that can be performed without specialized equipment, on virtually any scale. It has been explored to prepare conducting textiles functionalized by a self-doped poly(3,4-ethylenedioxythiophene) (PEDOT) derivative with methoxy-1-butane sulfonic acid side chains<sup>[7c,10]</sup> and PEDOT:polystyrene sulfonate (PEDOT:PSS),<sup>[4b,7a,b,11]</sup> with one of the highest reported bulk conductivities achieved in our previous work being  $14 \text{ S cm}^{-1}$ .<sup>[4b]</sup>

For e-textiles to be practically useful they must retain their electronic properties despite day-to-day wear as well as machine laundry. Encouragingly, several groups have reported water-washable polymer-based conducting textiles. Molina et al.<sup>[8c]</sup> in situ polymerized polypyrrole (PPy) onto a polyester fabric (counterion anthraquinone-2-sulfonic acid sodium salt (AQSA)), and noted that washing resulted in a loss of about one third of the counter-ions, and a 100% increase in resistance. Even more promising results are presented based on in situ chemical polymerization of PPy, polyaniline (PANI), and PEDOT (dopants:  $\text{FeCl}_3$ , ammonium persulfate (APS) and APS, respectively) onto silk fibers,<sup>[8e]</sup> in situ polymerization of PANI (dopant: APS) on cotton,<sup>[8d]</sup> PEDOT:PSS-dyed silk using dip

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coating,<sup>[4b]</sup> and PEDOT:PSS ink-jet printed onto a fabric<sup>[12]</sup>—all of which resulted in conducting textiles that could be machine washed several times with no or low (up to 10%) change in conductivity.

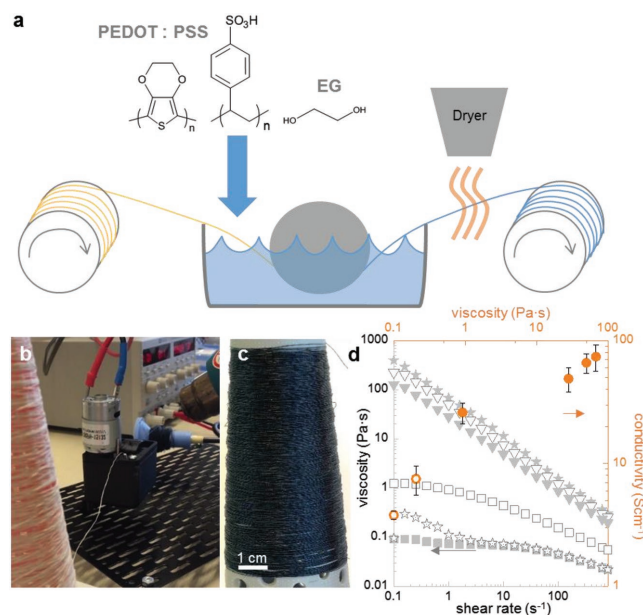
To assure availability of future e-textiles to the wide population, their manufacture should be adaptable to the processes currently available in the textile industry. Most studies on conducting textiles are based on batch processing and fall short of demonstrating that the materials can be produced and processed with large scale equipment. Moreover, we propose that the functionalization of yarns, rather than pieces of fabric, will provide a material suitable for patterning, which is a prerequisite for the construction of electrical devices and circuitry.<sup>[13]</sup> Patterned textiles can be fabricated by weaving, knitting, or stitching combinations of yarn into a fabric, as has been previously reported using metal-based yarns<sup>[3a,14]</sup> or manual techniques.<sup>[1a,c,2a,3a,4b,15]</sup> For large scale manufacture, these methods require the availability of a continuous length—at least 100 m—of conducting yarn.

Here, we demonstrate a continuous functionalization process for *Bombyx mori* silk yarns, reminiscent of the padding mangle commonly used for dyeing of fabrics in the textile industry. We develop a dye bath formulation where PEDOT:PSS constitutes the colorant, and use roll-to-roll (R2R) processing to dye over 100 m of silk yarns with a—for coated textiles—record high bulk conductivity of  $\approx 70 \text{ S cm}^{-1}$ . Our conducting yarns were further processed by machine embroidery to produce e-textile circuits, and by hand weaving to produce a versatile textile keyboard. Our e-textiles were machine laundered 15 times with no adverse effect on the electrical conductivity.

## 2. Results and Discussion

Silk is a natural protein fiber which is typically dyed using acid dyes.<sup>[16]</sup> An aqueous PEDOT:PSS dispersion, with its sulfonic acid groups and inherently low pH ( $\text{pH} \approx 2$ ), offers itself as a promising candidate for functional silk dyeing. A low pH ionizes the amino groups present in silk, and their combination with the negatively charged PSS can contribute to strong electrostatic interactions, resulting in a wash and wear-resistant functionalization of the silk. We have previously presented the successful dyeing of washable conducting silk yarns using a lab-scale dip-coating process.<sup>[4b]</sup> To apply this process to R2R dyeing, we here use a custom built device where a yarn continuously passes through a trough containing the dye bath, followed by drying and collection onto a take-up roller (Figure 1a,b). By this method, we are able to continuously dye any length of yarn. For further experiments we went on to dye 100 m of silk yarn with PEDOT:PSS (Figure 1c).

One drawback of continuous dyeing, compared to the batch process, is the limitation of the yarn's residence time in the dye bath. This reduces the time available for the water to swell the fiber, and for the dye to diffuse into the fiber. The effect was clearly seen in our first trials, where the as-received PEDOT:PSS dispersion (PH1000 from Heraeus) was used as dye bath. Decreasing the take-up rate from 1.7 to  $0.2 \text{ m min}^{-1}$  (residence time  $\approx 0.03$  and  $0.3 \text{ min}$  respectively) resulted in a doubled conductivity, i.e.,  $0.02 \text{ S cm}^{-1}$  instead of  $0.01 \text{ S cm}^{-1}$



**Figure 1.** a) Schematic representation of the setup for continuous dyeing: the pristine silk yarn (left) is pulled through a dye bath consisting of an aqueous PEDOT:PSS dispersion containing 5% ethylene glycol (EG), and dried with a heat gun when exiting the dye bath. b) Photograph of the continuous dyeing setup with the controlling voltage supply and c) photograph of a yarn bobbin with 100+ m of dyed yarn. d) The viscosity of the dye bath as a function of shear rate (left and bottom axes, grey symbols) for the as-received PEDOT:PSS dispersion (unfilled stars), PEDOT:PSS dispersion +5% EG (filled squares), and of the PEDOT:PSS+EG ink after sitting on a hot plate set to  $50^\circ\text{C}$  for 16.5 h (unfilled squares), 43 h (filled triangles), 46.5 h (unfilled triangles), and 47 h (filled stars). Electrical conductivity of the dyed yarns as a function of the dye bath viscosity at shear rate =  $1 \text{ s}^{-1}$  (right and top axes, orange open circles represent one layer dyed yarns, orange filled circles represent two layer dyed yarns, error bars represent the standard deviation based on ten measured samples).

(Table 1). The here reported values for conductivity refer to bulk conductivity ( $c$ ), calculated as in Equation (1)

$$c = \frac{l}{R \times A} \quad (1)$$

where  $l$  is the length of the measured yarn section,  $R$  is the resistance, and  $A$  is the cross-section area of the dyed yarn. The diameters of our dyed yarns are in the range of  $0.21\text{--}0.25 \text{ mm}$ , as measured by optical microscope (Table S1, Supporting Information).

To compensate for the short residence time, we increase the dye concentration gradient (with respect to pristine yarn) by removing water from the dye bath, thus increasing the absorption rate. The impact of concentrating the ink formulation can be measured as a change in both viscosity profile and in absolute viscosity (Figure 1d). The as-received PEDOT:PSS dispersion exhibits a nearly Newtonian profile, also after the addition of 5% ethylene glycol (EG). However, as the ink concentration increases, its viscosity profile becomes pseudo-plastic and its absolute viscosity increases over all shear rates tested. By comparing the viscosity of the different inks, at a selected shear rate of  $1 \text{ s}^{-1}$ , we find that there exists a direct correlation

**Table 1.** Dye bath formulations, processing parameters, and final conductivities of the yarns.

Dye formulation	No. of coating cycles	Take up rate [m min <sup>-1</sup> ]	Viscosity <sup>a)</sup>	Post treatment	Conductivity [S cm <sup>-1</sup> ]
PEDOT:PSS	1	1.7	0.1	none	0.01
PEDOT:PSS	1	0.2	0.1	none	0.02
PEDOT:PSS+EG	1	0.2	0.1	none	0.03
PEDOT:PSS+EG	1	0.2	0.1	EG	4 ± 1
PEDOT:PSS+EG	1	0.2	0.2	EG	8 ± 1
PEDOT:PSS+EG	1	0.2	0.2	DMSO	8 ± 2
PEDOT:PSS+EG	1	0.2	0.9	EG	11 ± 3
PEDOT:PSS+EG	1	0.2	0.9	DMSO	19 ± 3
PEDOT:PSS+EG	2	0.2	0.9	DMSO	26 ± 5
PEDOT:PSS+EG	2	0.2	28	DMSO	49 ± 11
PEDOT:PSS+EG	2	0.2	50	DMSO	66 ± 12
PEDOT:PSS+DMSO	2	0.2	53	DMSO	41 ± 7
PEDOT:PSS+EG	2	0.2	68	DMSO	74 ± 18

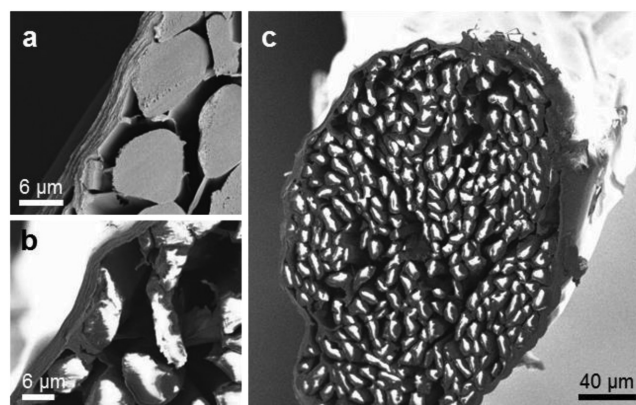
<sup>a)</sup>at shear rate = 1 s<sup>-1</sup>

between ink viscosity and final conductivity of the fibers (Table 1 and Figure 1d). In future experiments, increasing the physical length of the dye bath trough (here limited to about 6 cm) would allow for a higher take up rate without loss in residence time.

The electrical conductivity of PEDOT:PSS can be increased by several orders of magnitude by coprocessing and/or post treatment with high boiling solvents, such as dimethyl sulfoxide (DMSO) or ethylene glycol (EG) (e.g.<sup>[17]</sup>). We find that after an addition of 5% EG to our dye bath, the conductivity increases from 0.02 to 0.03 S cm<sup>-1</sup>. This relatively low effect on conductivity is presumably due to the short residence time in the dye bath, followed by quick drying by means of a heat gun. We therefore add a post-treatment step, where the dry dyed yarns are placed in a bath of EG or DMSO for 80 min, which resulted in a two orders of magnitude increase in yarn conductivity. By combining dye bath optimization, in terms of ink concentration, and post-treatments, and finally by increasing the amount of PEDOT:PSS in the dyed yarn by passing the yarn twice through the dye bath, we achieve a final electrical bulk conductivity of ≈70 S cm<sup>-1</sup> (Figure 1d and Table 1). These yarns were selected for use in continued experiments.

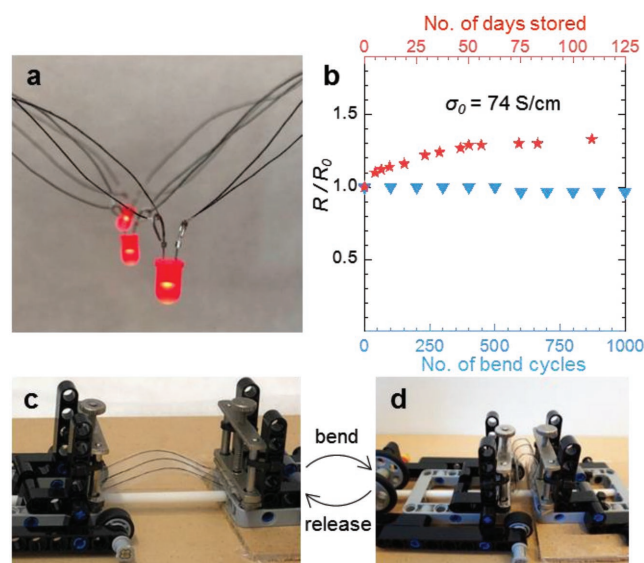
We further analyze our dyed conducting yarns by carrying out scanning electron microscopy (SEM) on their cross-section. Apparently the PEDOT:PSS-dye has solidified to form a film surrounding the individual silk fibers, and after the second dyeing cycle a thicker layer of coating is present around the yarn perimeter (Figure 2a). By also studying a cross-section of the yarn without pretreatment (i.e., sputtering), we find that the individual fibers show charging artefacts concentrated at their core. The lack of charging around the perimeter confirms that, in spite of the short dyeing time, conducting PEDOT:PSS has diffused 1–3 μm into the surface of individual silk fibers, resulting in a conducting outer layer (Figure 2b,c).

Our dyed silk yarns can be used electrical connectors, for example between a battery and several light-emitting diodes



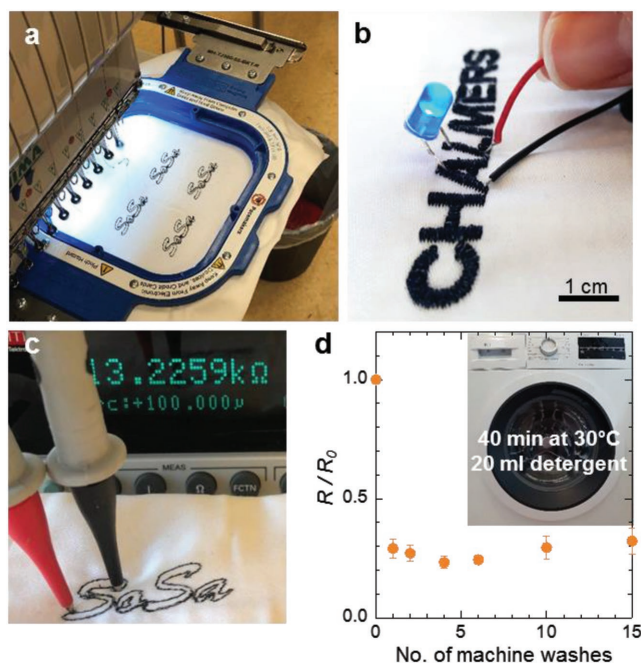
**Figure 2.** SEM-images of a) a close-up of the cross-section of the dyed silk yarn, sputtered with gold. b) A close-up of the nonsputtered cross-section of the dyed silk yarn, and c) SEM-image of the entire, nonsputtered, cross-section of the yarn.

(LEDs) (Figure 3a). The long-term properties of our yarns, stored under ambient conditions, were studied by repeated resistance measurements. We find that there is some increase, to around 1.3 times its original resistance, during the first month of storage, where after resistance levels off (Figure 3b). To further evaluate the resilience of our yarns, we constructed a LEGO-device to conduct repeated bending tests over a radius of 5 mm, and found that the electrical conductivity is not adversely affected, even after 1000 bend cycles (Figure 3b–d). The mechanical properties of the silk yarns: strain to break ≈10%, stress at break ≈130 MPa and Young's modulus ≈3 GPa, were retained after dyeing (Figure S1 and Table S1, Supporting Information).



**Figure 3.** a) The dyed silk yarns function as the electrical connector between several LEDs and a battery. b) The change in measured resistance as a function of bending (bottom x-axis, blue triangles) and storage under ambient conditions (top x-axis, red stars). Photographs of three pieces of dyed yarn mounted in the bending device during the c) release phase and, d) the bend phase.

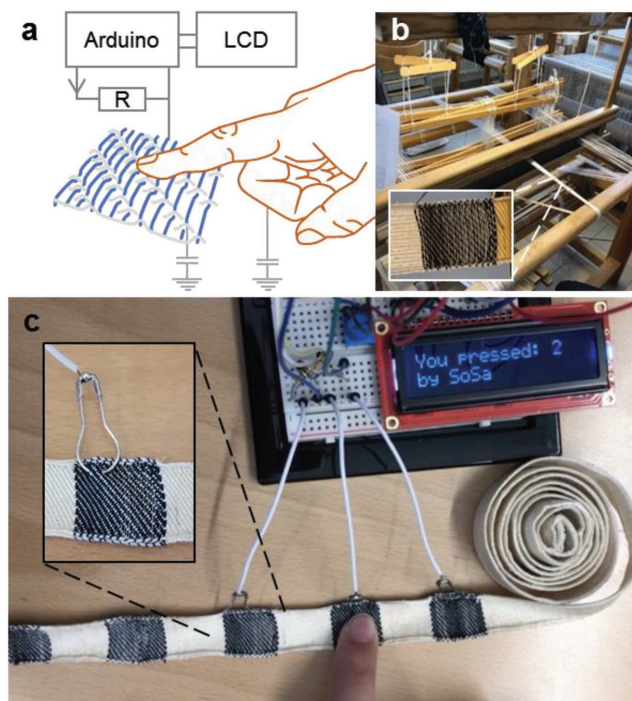




**Figure 4.** Photographs of a) machine embroidery using our dyed yarns, b) a machine embroidered text which functions as an electrical connector between an LED and a battery, and c) resistance measurement of an embroidered sample used in laundry tests. d) Average change in resistance versus the number of machine wash cycles (error bars represent the standard error, based on 12 measured samples).

To evaluate our yarns for use in traditional textile processes, we used an industrial type embroidery machine to design a variety of conducting patterns (Figure 4a,b; Movie S1, Supporting Information). By this stitching method, complex conducting patterns can be formed on any base fabric. Machine stitching involves repeatedly passing the yarns first through the small eye of a metal needle, and then through the fabric. Consequently, tensile strength in combination with a good resistance to abrasion will be required. Evidently, even after this abrasive process, our yarns are of high enough conductivity to function as a connector between an LED and a battery (Figure 4b). We used the embroidered textiles to evaluate the resistance of our conducting yarns to domestic laundering. Two pieces of fabric with a ten times repeated pattern ("SoSa") was machine laundered 15 times. Between wash cycles, after drying for at least 24 h, the electrical resistance along the length of the letter "S" was measured (Figure 4c) to evaluate the change in resistance. After the first washing cycle the average resistance dropped to a quarter of its original value (Figure 4d). Evidently the process of laundering has a "healing" effect on the dyed thread, which we rationalize by removal of excess PSS. After the first wash, the electrical resistance remains constant even after 15 machine wash cycles.

To produce a second e-textile device from our dyed yarns, we design an Arduino-based capacitive touch sensor (Figure 5a) connected to a liquid crystal display (LCD). The capacitive touch functionality is attained by polling the signal through a simple RC-circuit realized by a resistor and an open connector. The



**Figure 5.** a) Schematic representation of the capacitive touch sensor. b) A loom set up for band weaving of our dyed yarns, insert shows a length of the woven band. c) Photograph of the textile capacitive touch sensor with key no. 2 activated. The insert shows the safety pin electrical connection.

addition of an electrode, i.e., a large conducting surface such as a textile, to the open connector results in the formation of a capacitive path to ground. Upon touch, the user will constitute a parallel capacitive connection to ground, thereby changing the capacitance and the time-constant of the RC-circuit. We used a loom to construct a woven band of alternating conducting (dyed silk weft yarn) and insulating (nondyed silk weft yarn) areas (Figure 5b; Movie S2, Supporting Information). In weaving, two sets of yarns are interleaved: the warp yarns are suspended along the length of the loom, individually threaded through the dents of a reed to the take up roller. The motion of the reed, which pushes the weft yarn into the forming fabric, is aptly termed "beating-up:" yarns for weaving will need to possess a high tenacity, a recoverable extensibility of around 5%–10% and a high abrasion resistance. Our dyed yarns are sufficiently resilient for this process, and we produced a woven band that can serve as a versatile "sensors-on-a-roll" e-textile component for use, e.g., as a touch-sensitive textile keyboard. We connected the textile to a conventional electronics breadboard, using safety pins soldered to a thin copper wire. The software responds to the change in capacitance by displaying a message on the LCD, communicating the selected "key" (Figure 5c; Movie S3, Supporting Information).

### 3. Conclusions

Over 100 m of electrically conducting silk yarn was manufactured by a roll-to-roll dyeing process using a

PEDOT:PSS-based aqueous dye bath. Contrary to many other upscaling studies, we obtain yarns with improved conductivity of about  $70 \text{ S cm}^{-1}$ , a record high result for coated fibers and yarns.<sup>[6b]</sup> Our dyed yarn retains its conducting properties after repeated bending, industrial embroidery as well as 15 cycles of domestic laundering. Finally, we used hand weaving to produce a fully textile keyboard with capacitive touch functionality.

## 4. Experimental Section

**Materials:** Degummed silk sewing thread was purchased from Aurora Silk. The dyeing process included a prewash in a 1:50 (yarn:water) wash bath using  $4 \text{ g dm}^{-3}$  (with respect to water) silk detergent (Zenit) and  $4 \text{ g dm}^{-3}$  ammonia (25%, Merck Millipore). The dye bath contained PH1000 PEDOT:PSS dispersion (1.1–1.3 wt% solid as-received, Heraeus) and 5 vol% of ethylene glycol (EG) or dimethyl sulfoxide (DMSO) (Sigma-Aldrich), and the post treatment used EG or DMSO. Water was removed from the dye bath by evaporation on a  $50^\circ\text{C}$  hotplate for 2.5–47 h (cf. Table S1, Supporting Information).

**Machine Embroidery:** Embroidery was carried out on an industrial automatic embroidery machine, Tajima TMBP-S1501C, where the thread was embroidered onto a cotton fabric at 600–700 stitches per min.

**Machine Washing:** The embroidered fabric was laundered in a domestic washing machine (Bosch) using the wool wash program (40 min at  $30^\circ\text{C}$  and 800 rpm spin), with 20 mL of commercial detergent (Neutral) containing 5%–15% anionic and nonionic surfactants, and <5% enzymes.

**Weaving and Capacitive Sensing:** Weaving was carried out in a hand loom with a reed of 9 dents  $\text{cm}^{-1}$  and two threads per dent, using mercerized cotton ( $16\,000 \text{ m kg}^{-1}$ ) as warp yarn. The weave structure was a weft-faced twill, and the final woven band was 2 cm by 1.5 m, with alternating 2 cm long conducting and nonconducting regions. The woven band was connected via safety pins to an UNO microcontroller (Arduino) and an LCD (Sparkfun Electronics).

**Electrical Characterization:** The resistance was measured using a Keithley 2400 sourcemeter in 2-point configuration. For characterization of electrical conductivity, the dyed silk yarn was placed on a glass slide and contacted by applying silver paint (Agar Scientific) on the yarn surface. The yarn diameter was measured by optical microscopy. The resistance of washed samples was measured by placing two probes directly on the dyed silk yarn. Bending resilience was characterized using an in-house built LEGO device (Lego System A/S), which repeatedly bent the yarns to a diameter of 5 mm. The resistance was measured after 100 bending cycles, a process which was repeated ten times.

**Tensile Testing:** Tensile testing was carried out using an Instron tensile tester 5565A, with a gauge length of 5 cm and a drawing speed  $10 \text{ mm min}^{-1}$ .

**SEM:** Scanning electron microscopy (SEM) images were taken at an acceleration voltage of 3 kV using a Leo Ultra 55 SEM equipped with a field emission gun and a secondary electron detector. The yarn was frozen in liquid nitrogen prior to cutting. The samples were either sputtered with gold or unsputtered.

**Rheology:** The viscosity was measured as a function of shear rates ( $0.1\text{--}800 \text{ s}^{-1}$ ) using a DHR-3 (TA Instruments) with a cone and plate geometry having a gap of  $26 \mu\text{m}$ . The cone diameter was 40 mm and cone angle was  $1^\circ$ . The viscosity of all dispersions was determined at  $25^\circ\text{C}$ .

## Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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## Conflict of Interest

The authors declare no conflict of interest.

## Keywords

conducting fibers, electronic textile, e-textile, PEDOT:PSS

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